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A high-resolution air chemistry record from an Alpine ice core: Fiescherhorn glacier, Swiss Alps

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Abstract. Glaciochemical studies at midlatitudes promise to contribute significantly to the understanding of the atmospheric cycling of species with short atmospheric lifetimes. Here we present results of chemical analyses of environmentally relevant species performed on an ice core from Fiescherhorn glacier, Swiss Alps (3890 m above sea level). This glacier site is unique since it is located near the high-alpine research station Jungfraujoch. There long-term meteorological and air quality measurements exist, which were used to calibrate the paleodata. The 77-m-long ice core was dated by annual layer counting using the seasonally varying signals of tritium and $\delta^{18}O$. It covers the time period 1946-1988 and shows a high net accumulation of water of 1.4 m yr^{-1} allowing for the reconstruction of high-resolution environmental records. Chemical composition was dominated by secondary aerosol constituents as well as mineral dust components, characterizing the Fiescherhorn site as a relatively unpolluted continental site. Concentrations of species like ammonium, nitrate, and sulfate showed an increasing trend from 1946 until about 1975, reflecting anthropogenic emission trends in western Europe. For mineral dust tracers, no trends were obvious, whereas chloride and sodium showed slightly higher levels from 1965 until 1988, indicating a change in the strength of sea-salt transport. Good agreement between the sulfate paleorecord with direct atmospheric measurements was found (correlation coefficient r^2=0.41). Thus a "calibration" of the paleorecord over a significant period of time could be conducted, revealing an average scavenging ratio of 180 for sulfate.

1. Introduction

Ice cores from polar ice sheets have now become established as important archives for paleoclimatic and paleoatmospheric information covering timescales of up to several hundred millennia (see, e.g., the special issue Greenland Summit Ice Cores GISP2/GRIP in *Journal of Geophysical Research, 102(C12)*, 26,315-26,886, 1997). Much less attention has been paid to the potential of alpine glaciers to provide environmental records since the characteristics of such glaciers, namely limited size, location in a complex topography, mostly unknown flow patterns, were believed to hinder or even prevent interpretation. In addition, suitable glaciers are often small in the horizontal and vertical extension, thus limiting the accessible timescales to, at best, several thousands of years in most cases. Nevertheless, glaciochemical studies at midlatitudes promise to contribute significantly to the understanding of atmospheric cycles [e.g., *Thompson et al.*, 1995, 1997], particularly of species with short atmospheric lifetimes of the order of days. Such species are nonuniformly distributed in the atmosphere, with highest concentrations close to the emission sources. A good example are atmospheric aerosol particles, which have attracted attention in climate research recently since their cooling effect on the atmosphere might partly compensate for the warming by anthropogenically emitted greenhouse gases [*Charlson et al.*, 1991]. In order to quantify the effect of anthropogenic aerosols, reliable concentration data sets from the source regions are necessary.

For the purposes of reconstructing atmospheric concentrations of short-lived species and to document the effects of anthropogenic emissions on air pollution, the European Alps are well suited, since they are surrounded by highly populated and industrialized areas. Owing to their elevation, they are partly covered by glaciers. Since these glaciers are located at high altitudes, the transport of air from the emission sources is hindered by atmospheric stability [*Baltensperger et al.*, 1997]. Thus they experience mainly relatively unpolluted, continental air [*Wagenbach*, 1997]. Nevertheless, the proximity to the emission sources facilitates the interpretation of the observed records since the atmospheric transport patterns are known [*Kromp-Kolb et al.*, 1997; *Lugauer et al.*, 1998]. Furthermore, historical meteorological and air quality measurements exist, which can be used to calibrate the paleo(climatic) data [*Gehrig*, 1996; *Kromp-Kolb et al.*, 1993]. This combination of suitable glacier archives with long-term, high-quality instrumental and air chemistry records available in the vicinity is unique. Only at the South Pole does a comparable situation exist with continuous measurements of a physical aerosol parameter, the scattering coefficient, since 1979. This physical parameter was related to firn core chemistry over a 10-year period [*Bergin et al.*, 1998], since no direct air chemistry data were collected. For Europe, in addition to instrumental records, emission estimates for environmentally relevant species such as sulfur dioxide, nitrogen oxides, ammonia, and heavy metals are available over time [*Mylona*, 1993; *Asman et al.*, 1988]. Since precipitation rates are high in

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the Alps, records with high time resolution can be obtained, another strength of Alpine ice core investigations.

In the Alps, ice core work has so far been focused mainly on the Monte Rosa area in Switzerland and the Mont Blanc area in France. In 1976, at Colle Gnifetti, a glacier saddle at 4450 m above sea level (asl) in the Monte Rosa massif, two cores were drilled in order to study glaciological parameters, to reveal the snow accumulation rate, and to determine the history of atmospheric trace components [*Oeschger et al.*, 1977]. Meanwhile, various records of chemical trace species and mineral dust have been published from different cores reaching bedrock, clearly demonstrating the impact of anthropogenic emissions on the snow composition [*Haeberli et al.*, 1983; *Gäggeler et al.*, 1983; *Schotterer et al.*, 1985; *Wagenbach et al.*, 1988; *Wagenbach and Geis*, 1989; *Döscher et al.*, 1995a, 1996; *Wagenbach et al.*, 1996]. However, the Colle Gnifetti experiences strong winds, especially in winter, eroding significant parts of the snow accumulation. Thus records from this site represent mainly summer conditions. Nevertheless, a comparison of the fluxes of major ions at Colle Gnifetti and Col du Dome (4250 m asl), located below the Mont Blanc summit, over a 3.5-year period showed excellent agreement and revealed the Col du Dome as a site with high accumulation and seasonal resolution [*Maupetit et al.*, 1995].

The limited number of ice core studies conducted in the Alps so far is partly due to the fact that only a few potential drilling sites exist. Using altitude and surface as well as bed geometry as main criteria to select suitable glacier sites, only four high-elevation areas have been identified (Monte Rosa, Bernese Alps, Grand Combin, and Mont Blanc [*Funk*, 1994]; see Figure 1 for locations). Here we present data from an ice core drilled at the Fiescherhorn glacier, which is located in one of these areas (Bernese Alps, Switzerland). This glacier is probably the only suitable drilling site on the northern main Alpine chain. The site was chosen with the expectation of higher accumulation rates than those observed on wind-exposed glacier saddles such as the Colle Gnifetti, promising records with a resolution of several data points per year, and also because of the opportunity to link the ice core data to various instrumental records from the nearby high-alpine research station Jungfraujoch located at 3450 m asl, which is only 6 km horizontal distance to the west. Besides, it was assumed to represent the conditions of the catchment area north and northwest of the Alps, where major source regions for air pollution are situated (Germany, Benelux countries, France; see Figure 1). The initial purpose for drilling was the reconstruction of the Chernobyl fallout in May 1986 [*Gäggeler et al.*, 1986; *Haeberli et al.*, 1988].

2. Characteristics and Location of Fiescherhorn Glacier

The drilling site chosen for this study forms a part of the accumulation area of the Fiescherhorn glacier (8°02'46"E, 46°32'53"N) (Figure 2). This area is located on a plateau with a size of about 0.5 km^2 between the mountains Grossfiescherhorn (4049 m asl), Hinterfiescherhorn (4025 m asl), and Ochs (3900 m asl). The plateau shows a slope of 16% toward the east and a mean elevation of about 3880 m asl. Radar soundings and a first test drilling performed in 1986 revealed a glacier thickness of about 70 m and a temperature of -5.2 ± 0.5°C at 7.5 m below the surface (J. Schweizer, unpublished data, 1986). Thus this part of the glacier belongs to the cold infiltration zone, where meltwater percolation is either absent or meltwater immediately refreezes some centimetres below the surface when it has formed at the surface under the influence of solar radiation. This fact was confirmed by visual inspection of the firn part of the core, showing only thin layers of ice lenses, as well as by the regular seasonality of the tritium concentration and the $\delta^{18}O$ isotopic ratio (see below). A disturbance of underlying annual layers can therefore be neglected, which is a prerequiste for interpreting concentrations records from the ice core. Assuming a mean temperature gradient of 0.016 °C m^{-1}, a corresponding temperature of about -4°C at bedrock is obtained, indicating that the glacier is frozen to the bedrock (J. Schweizer, unpublished data, 1986). The final deep drilling of 77 m took place in 1989 at an elevation of 3890 m asl, where the radar soundings suggested a less steep bedrock and a glacier thickness of about 85 m [*Stampfli*, 1989].

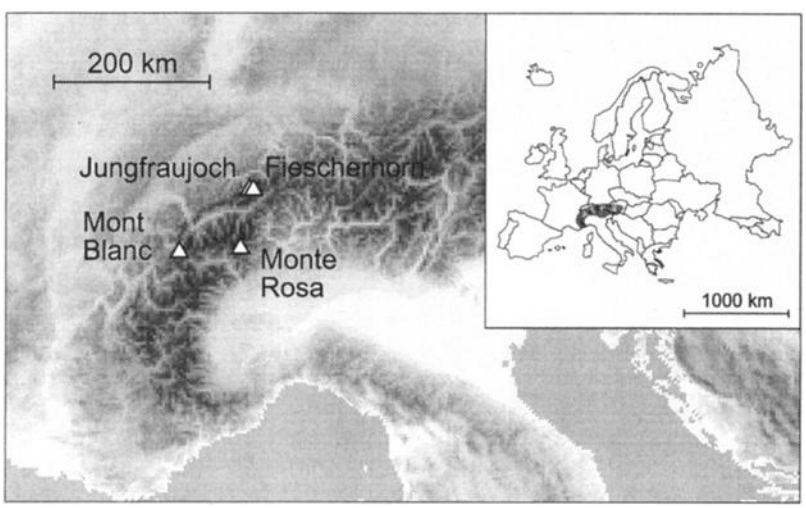

Figure 1. Topographic map of the Alps showing the positions of the Jungfraujoch and the Fiescherhorn glacier in the Bernese Alps, as well as of the Mont Blanc and the Monte Rosa area. The Col du Dome (not marked) is located below the summit of the Mont Blanc, and the Grand Combin (not marked) is close to the Monte Rosa. Inset shows a map of Europe where the Alps are indicated.

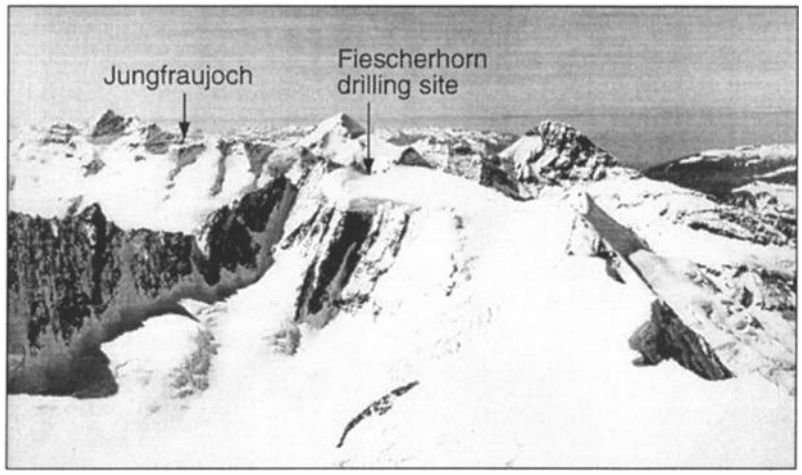

Figure 2. View of the Fiescherhorn glacier from the southeast with the position of the drilling in 1989. In the background are the mountains Mönch, Eiger, and Jungfrau (from right) with the high-alpine research station Jungfraujoch (indicated). Photo reproduced with permission of W. P. Burkhardt, 1998.

3. Drilling, Sample Handling, Chemical Analysis, and Dating of the Core

The 77-m-long ice core with a diameter of 7.5 cm was electromechanically drilled. Ice core sections 0.5 to 0.9 m long were recovered, sealed at the drilling site in polyethylene bags, and kept frozen at -30°C until the processing started. In a cold room, the core sections were decontaminated by mechanically removing 0.5 cm of the outer part of the core and cut into 4- to 23-cm-long samples using a band saw. The length of the samples was chosen in order to assure a frequency of at least 12 to 15 samples per assumed year. The samples were melted only prior to the analyses. All materials assumed to come in contact with the samples (e.g., tubes and vials) had been carefully precleaned with ultrapure water (18 MΩ cm quality) in the laboratory [*Schwikowski et al.*, 1997]. Concentrations of the main water-soluble aerosol species Ca^{2+}, Cl^-, K^+, Mg^{2+}, Na^+, NH_4^+, NO_3^-, and SO_4^{2-} were determined by ion chromatography with conductivity detection [*Döscher et al.*, 1995b; *Schwikowski*, 1997]. Analytical detection limits and blank values obtained by analyzing artificial ice cores are given in Table 1. The artificial ice cores were made by freezing of ultrapure water (18 MΩ cm quality) in the polyethylene bags normally used for storing and were prepared identically to the real ice cores. The detection limits were controlled by the blank values and not by the sensitivity of the ion chromatography. Therefore concentrations measured in the actual ice samples were corrected for the blank values. Concentrations of tritium as well as the stable isotopic ratios of water δD and $\delta^{18}O$ were determined in the same samples. For details regarding the tritium analyses, see *Schotterer et al.* [1998].

Dating of the ice core was performed by annual layer counting using the seasonally varying signals of the concentration of tritium and of the isotopic ratio $\delta^{18}O$. Low and high concentrations of tritium as well as low and high $\delta^{18}O$ values represent winter snow and summer snow, respectively. In the case of tritium, the seasonality is superimposed by a concentration trend showing a maximum in the year 1963 with highest release of tritium into the atmosphere by atomic bomb testing. The annual layer counting was therefore adjusted to the 1963 horizon as a reference point [*Schotterer et al.*, 1998]. As an additional reference horizon, the Saharan dust event of 1977 [*Oeschger et al.*, 1977; *Haeberli et al.*, 1983; *Schotterer et al.*, 1985; *Wagenbach and Geis*, 1989; *Döscher et al.*, 1996], identified by high concentrations of Ca^{2+}, was used. The suitability of this dating method is illustrated in Figure 3, where the seasonality of the concentration of tritium becomes obvious. A similarly pronounced seasonality is visible in the $\delta^{18}O$ record [see also *Schotterer et al.*, 1997]. By using a combination of both T and $\delta^{18}O$, a timescale with an uncertainty of only ±1 year could be deduced.

4. Results and Discussion

4.1. Concentration Records and Main Atmospheric Sources of Chemical Trace Species

Dating of the 77-m-long ice core revealed that it covers the time period 1946-1988. Thus the Fiescherhorn glacier is indeed a glacier with a high net accumulation, which varies between 1 and 2 m water equivalent per year with an annual mean value of 1.4 m water equivalent [see also *Schotterer et al.*, 1997]. However, this is only half of the precipitation obtained from measurements in the Jungfrau region at elevations from 3000 to 3500 m [*Barry*, 1992]. Even if taking the difficulties of precipitation measurements in high mountains into consideration, it cannot totally be excluded that precipitation at Fiescherhorn is only

Table 1. Detection Limits of Ion Chromatography (IC) and Blank Values of Different Ionic Species

	Cl^-	NO_3^-	SO_4^{2-}	Ca^{2+}	K^+	Mg^{2+}	Na^+	NH_4^+
Detection limits of IC, μg L^{-1}	5.0	2.0	5.0	10.0	3.2	1.5	1.6	0.6
Blank values, μg L^{-1}	11.4	2.2	5.5	11.4	4.0	1.8	7.5	0.6

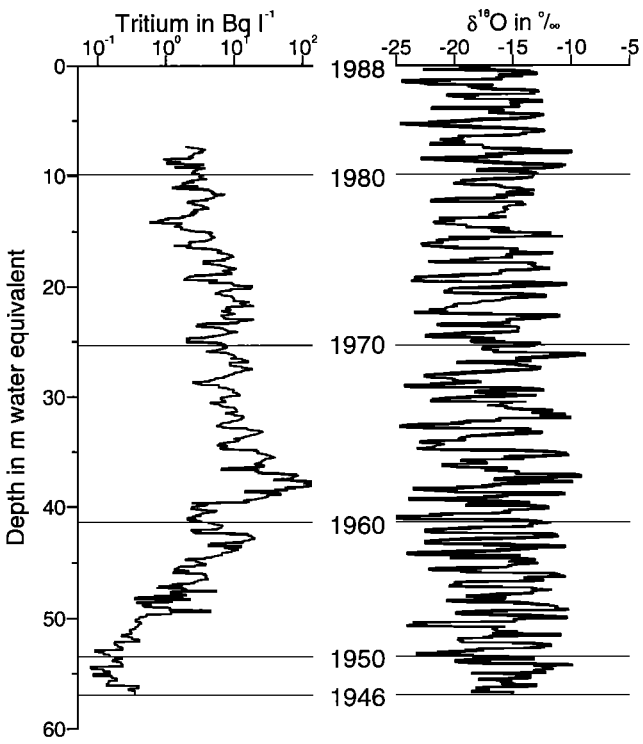

Figure 3. Variations of the concentrations of tritium and of $\delta^{18}O$ along the ice core. Note that the tritium concentration is plotted on a logarithmic scale and that samples from the topmost 7 m are not analyzed so far. The depth of the ice core of 57 m water equivalent represents the total length of 77 m.

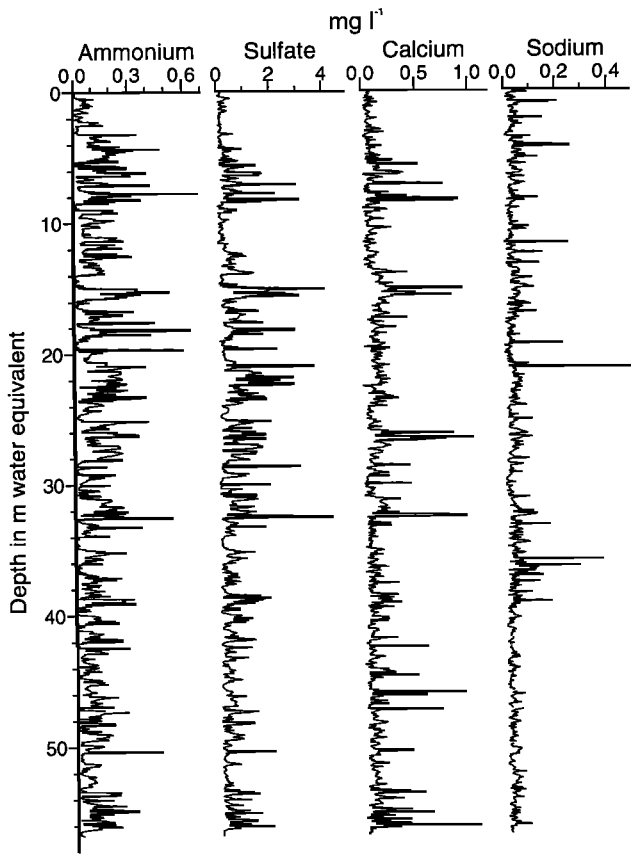

Figure 4a. High resolution concentration records of NH_4^+, SO_4^{2-}, Ca^{2+}, and Na^+ versus depth of the ice core in meters water equivalent.

partly preserved. In this case it can still be assumed that a representative portion of the precipitation is preserved, since the average $\delta^{18}O$ value of $-17^0/_{00}$, which is extremely sensitive to any disturbance or loss, fits well with the $\delta^{18}O$/altitude relationship obtained from the Swiss network for isotopes in precipitation [*Schotterer et al.*, 1997]. The high net accumulation allows one to obtain high-resolution concentration records. In order to illustrate the characteristics of the concentration records, they are displayed versus depth in meters water equivalent in Figures 4a and 4b. They consist of a total of 831 samples, with 9 to 34 samples per year, depending on the actual accumulation. From the records a few characteristics are evident. (1) Large fluctuations of the concentrations of the investigated ionic species are observed. (2) The fluctuations of, for example, ammonium and sulfate are related, whereas those of calcium and sodium follow a different pattern. (3) For some species such as ammonium and sulfate the fluctuations are superimposed by a trend in the concentrations. These characteristics are discussed in detail in the following.

Generally, concentrations of all species determined are lognormally distributed (Figure 4c). Median values varied between 0.009 and 0.324 mg L^{-1}, depending on the species and are given in Table 2. The overall chemical composition is dominated by the ions SO_4^{2-}, Ca^{2+}, NH_4^+, and NO_3^-, arranged in descending values of the median concentrations on an equivalent basis (Table 3). The sum of anions and cations are balanced within 10% (Table 3). However, one has to bear in mind that neither H^+ nor HCO_3^- was analyzed, and both can be present in snow samples [*Ronseaux and Delmas*, 1988; *Maupetit and Delmas*, 1994]. Thus the snow chemistry at the Fiescherhorn glacier is characterized by constituents originating mainly from

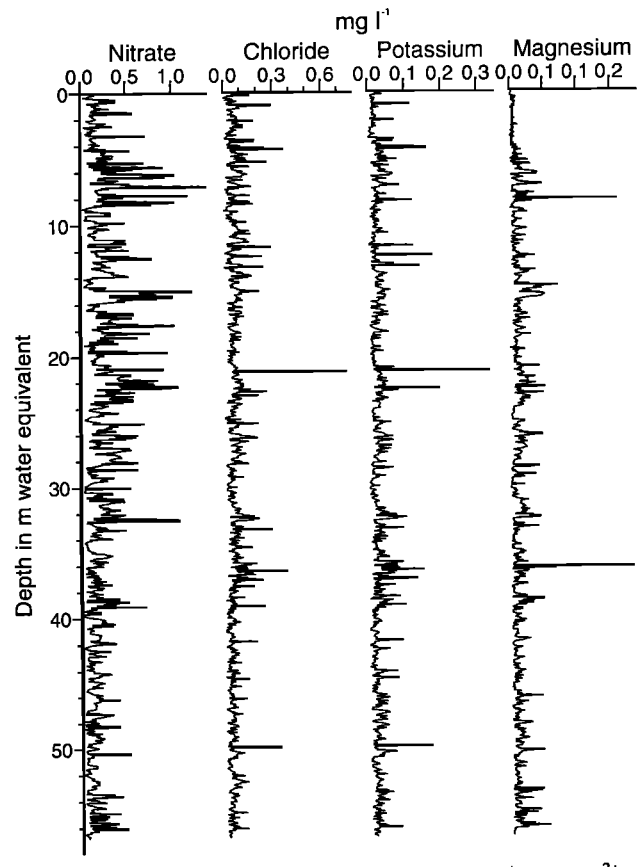

Figure 4b. Same as Figure 4a, but for NO_3^-, Cl^-, K^+, and Mg^{2+}.

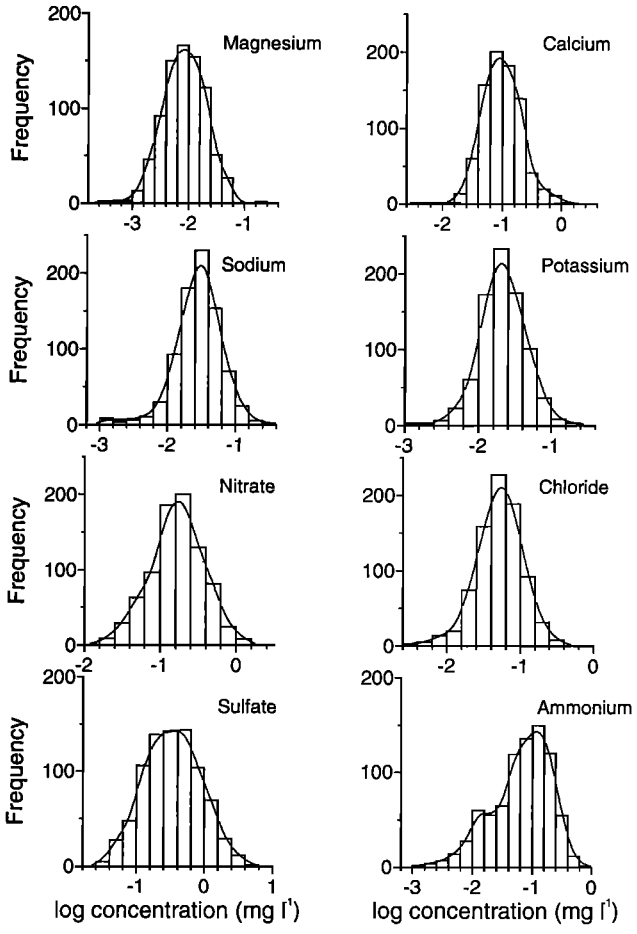

Figure 4c. Frequency distribution of the logarithmic values of the concentrations of various ionic species. To guide the eye, spline fits are shown (thin curves).

anthropogenically derived secondary aerosols and primary mineral dust particles of natural or anthropogenic origin. This is corroborated by a correlation analysis revealing high correlation coefficients r^2 between SO_4^{2-}, NO_3^-, and NH_4^+, on one hand, and Ca^{2+} and Mg^{2+} on the other hand (Table 4). A similarly high correlation coefficient r^2 was observed for Cl^- and Na^+ (r^2=0.66, Table 4), pointing to sea salt as a dominating source for these two species. This is corroborated by a Cl^-/Na^+ concentration ratio of 1.14 (molar ratio), which is close to the ratios measured in surface samples of bulk seawater of 1.16-1.17 [*Keene et al.*, 1986]. However, the sea-salt contribution to the overall chemical inventory is minor (about 12%, Table 3). Interestingly, K^+ shows the best correlation with the two sea-salt components Cl^- and Na^+, although the K^+/Na^+ concentration ratio of 0.32 significantly deviates from the sea-salt ratio (0.022, molar ratio [*Keene et al.*, 1986]), suggesting that only 5% of K^+ originated from sea salt. The main source of K^+ seems to be mineral dust, with obviously varying K^+/Ca^{2+} ratios. The same phenomenon was previously observed for snow samples from the French Alps [*Maupetit and Delmas*, 1994]; they attributed the correlation with sea salt components to the effect of simultaneous deposition. Ca^{2+} and Mg^{2+} correlate not only with each other but also with SO_4^{2-}, indicating that SO_4^{2-} is partly associated with mineral dust particles. Taking the mean pre-industrial SO_4^{2-}/Ca^{2+} concentration ratio of 0.18 (molar ratio) obtained for the time period 1756-1795 from an ice core drilled on Colle Gnifetti, Monte Rosa (M. Schwikowski et al., manuscript in preparation, 1999), 16% of the SO_4^{2-} on Fiescherhorn is of mineral dust, i.e., natural origin.

The median concentrations are in good agreement with data from the Colle Gnifetti (4450 m asl, Swiss Alps) where concentrations of 0.049 mg L^{-1} for Cl^-, 0.16 mg L^{-1} for NO_3^-, 0.45 mg L^{-1} for SO_4^{2-} (time period 1965-1981 [*Döscher et al.*, 1995b]) and about 0.12 mg L^{-1} for NH_4^+ (time period 1950-1981 [*Döscher et al.*, 1996]) were found. This is unexpected, since the Colle Gnifetti accumulates mainly summer snow, whereas winter snow is eroded by wind. Considering the seasonality of the trace species concentrations in high-alpine snow as explained below, lower concentrations at Fiescherhorn, where precipitation during all seasons is preserved, would be expected. However, the effect of different accumulation patterns at both glacier sites might be compensated by the lower elevation of the Fiescherhorn, since the atmospheric aerosol concentrations decrease with height. This was confirmed by continuous measurements over a 1.5-year period showing a 40% lower aerosol surface area concentration at the Colle Gnifetti than at the Jungfraujoch, with a height difference of about 1000 m between both sites [*Lugauer et al.*, 1998]. In order to compare our data with results obtained from a 13-m firn core from Col du Dome (4250 m asl, Mont Blanc summit area, northern French Alps) covering the years 1987-1990 [*Maupetit et al.*, 1995], averages were calculated for the period 1987-1988. Col du Dome is also a high accumulating glacier site with a net accumulation rate varying between 0.8 and 1.6 m water equivalent per year. Interestingly, concentrations in firn at Col du Dome are significantly enhanced for most ionic species compared to Fiescherhorn (factors of 1.6, 2.8, 2.8, 2.6, and 2.2 for NO_3^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, and NH_4^+, respectively) except for Na^+, Cl^-, and K^+ (factors of 0.5, 0.6, and 0.9, respectively). This difference seems to be mainly due to high concentrations in summer snow at Col du Dome (NO_3^- 0.646; SO_4^{2-} 1.113; Ca^{2+} 0.136; Mg^{2+} 0.014; and NH_4^+ 0.288 mg L^{-1} [*Maupetit et al.*, 1995]), possibly caused by a local pollution

Table 2. Median Concentrations of Major Ionic Species of Winter Snow, Summer Snow, and All Samples, Along With Summer to Winter Ratios

	n	$\delta^{18}O$, ‰	Cl^-, mg L^{-1}	NO_3^-, mg L^{-1}	SO_4^{2-}, mg L^{-1}	Ca^{2+}, mg L^{-1}	K^+, mg L^{-1}	Mg^{2+}, mg L^{-1}	Na^+, mg L^{-1}	NH_4^+, mg L^{-1}
All	831	-16.22	0.053	0.167	0.324	0.096	0.021	0.009	0.029	0.080
Winter	230	-20.87	0.050	0.135	0.253	0.099	0.018	0.007	0.030	0.029
Summer	298	-13.17	0.058	0.210	0.438	0.108	0.024	0.011	0.031	0.128
Ratio			1.16	1.56	1.73	1.09	1.33	1.57	1.03	4.41

Winter snow is defined as snow with $\delta^{18}O \leq -19‰$, and summer snow is defined as snow with $\delta^{18}O > -15‰$. Here n is number of samples.

Table 3. Ion Balance of Median Concentrations of All Samples

	Median, μeq. L^{-1}	Percentage of Measured Samples, %
	Anions	
Cl^-	1.51	14
NO_3^-	2.69	25
SO_4^{2-}	6.75	61
Total	10.95	
	Cations	
Ca^{2+}	4.8	40.5
K^+	0.5	4
Mg^{2+}	0.8	7
Na^+	1.3	11
NH_4^+	4.44	37.5
Total	11.84	

source [*Maupetit and Delmas,* 1994]. Nevertheless, the general agreement of the chemical composition of snow and ice from various high-elevation glacier sites in the Alps gives confidence that these glaciers are suitable as archives for the reconstruction of the European background atmosphere, since local effects seem to be negligible, at least for the considered species. Furthermore, the different data sets exhibit a common chemical signature of high-alpine precipitation with strong contributions of anthropogenically derived species as well as mineral dust constituents and a minor input of sea salt tracers.

4.2. Seasonal Variations

In the case of ammonium and, to a lesser extent, of nitrate and sulfate, the large fluctuations of the concentrations show seasonalities corresponding to those of tritium and $\delta^{18}O$ (Figure 5). As explained above, lowest values of tritium are typical for snow formed in winter when the exchange between the stratospheric reservoir of thermonuclear tritium and the troposphere is limited [*Schotterer et al.,* 1998]. The main injection occurs in spring and early summer. In winter the $\delta^{18}O$ also shows the most negative values because the temperatures during snow formation are lowest. The aerosol-borne chemical species NH_4^+, NO_3^-, and SO_4^{2-} are produced via gas-to-particle conversion of the gaseous precursors NH_3, NO_X, and SO_2. This production takes place mainly in the atmospheric boundary layer (ABL), where the emission sources of the precursors are located. In winter, high-alpine sites are decoupled from the ABL owing to the strong stability of the atmosphere, as demonstrated by continuous aerosol monitoring performed at high-elevation sites above 3100 m asl [*Baltensperger et al.,* 1997; *Lugauer et al.,* 1998]. This decoupling results in very low concentrations in snow of those species originating in the ABL. In summer, vertical transport of ABL air, even up to the highest summits in the Alps, occurs owing to convection, reflected in enhanced concentrations of ABL-originating species in the precipitating snow. In order to quantify this effect, the ice core samples were attributed to winter and summer snow using the $\delta^{18}O$ as indicator. On the basis of a $\delta^{18}O$/temperature relation for event-based samples from Jungfraujoch and equally sliced ice core segments from Fiescherhorn for the years 1983/1984 [*Schotterer et al.,* 1997], winter snow (months December, January, February, and March) was defined as snow with $\delta^{18}O \leq -19^0/_{00}$ and summer snow was defined as that with $\delta^{18}O > -15^0/_{00}$ (months June, July, August, and September). The corresponding median concentrations as well as the summer to winter ratios for the different species are summarized in Table 2. The seasonality is most pronounced for NH_4^+ with a ratio of 4.4, which was also observed at the Colle Gnifetti, where it was explained by a superimposition of atmospheric transport and emissions of NH_3 from agricultural activities, both maximal in summer [*Döscher et al.,* 1996]. A closer look at the annual summer to winter ratios revealed a strong year-to-year variability with a high correlation between the ratios of individual species ($r^2 = 0.79$ for NO_3^- and SO_4^{2-}, 0.57 for NO_3^- and NH_4^+, 0.61 for NH_4^+ and SO_4^{2-}). The correlation indicates that either a changing seasonality of precipitation or a changing intensity of convective transport affects the ratios rather than varying source strengths. The observed seasonality is consistent with results from an alpine wide snow sampling program carried out in the years from 1991 to 1993, focusing on the chemical composition of the high alpine winter and spring snow pack, although only spring to winter ratios are available from this study (High alpine snow pack chemistry study SNOSP [*Nickus et al.,* 1997]). For the sites in the western part of the Alps these ratios amounted to 4.0, 2.1, and 2.7 for NH_4^+, NO_3^-, and SO_4^{2-}, respectively, with spring comprising the months March to May and winter the months October to February. The seasonality of the concentrations was less distinct for the sites in the eastern Alps, reflecting most likely the generally lower elevation. In addition, the data set revealed a remarkable year-to-year variation of the seasonality. Continuous measurements of aerosol SO_4^{2-} at the Jungfraujoch [*Gehrig,* 1996] (see below) showed a summer to winter ratio of 1.85 for the period 1973 to 1988, which agrees well with the Fiescherhorn ratio. A stronger seasonal cycle was observed for the concentrations of particulate NH_4^+, NO_3^-, and SO_4^{2-} in air by continuous analysis performed over 1 year at another high-elevation mountain site, the Sonnblick Observatory in the Austrian Alps [*Kasper and Puxbaum,* 1994]. Summer

Table 4. Ion Correlation Matrix for the 831 Samples

	NH_4^+	NO_3^-	SO_4^{2-}	Na^+	Cl^-	Ca^{2+}	Mg^{2+}	K^+
NH_4^+	-	0.64	0.55	0.13	0.24	0.19	0.36	0.22
NO_3^-		-	0.74	0.19	0.32	0.28	0.42	0.25
SO_4^{2-}			-	0.18	0.21	0.40	0.58	0.29
Na^+				-	0.66	0.32	0.33	0.69
Cl^-					-	0.28	0.36	0.59
Ca^{2+}						-	0.61	0.35
Mg^{2+}							-	0.44
K^+								-

Given are correlation coefficients r^2 calculated from the logarithmic concentration values.

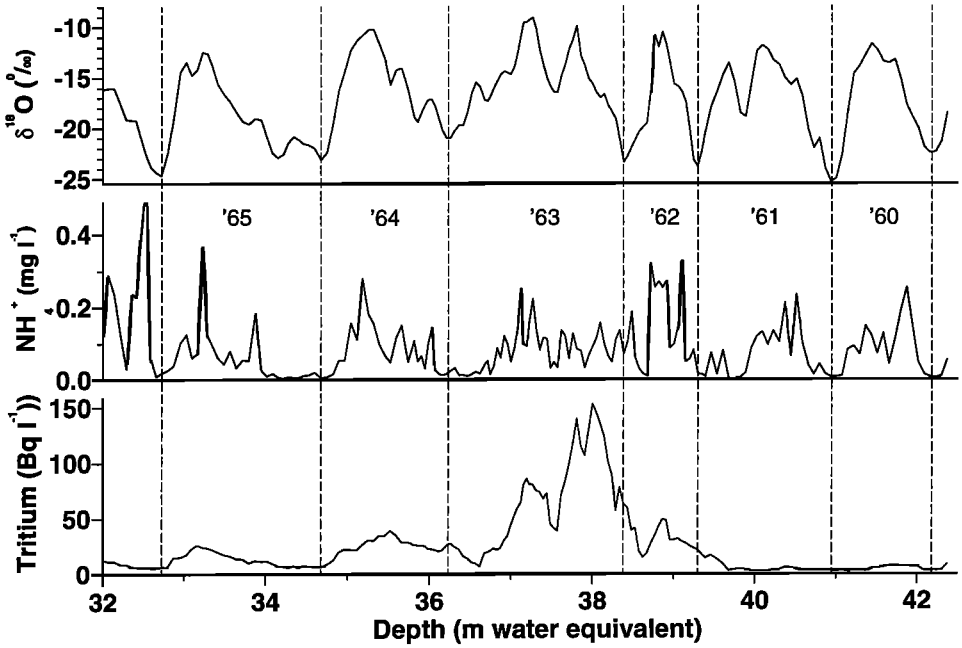

Figure 5. Magnification of a selected section of the records of tritium, NH_4^+, and $\delta^{18}O$, all showing a parallel seasonality. The dating procedure by counting individual years is indicated.

(June-September) to winter (December-March) ratios of 5, 4, and 4 were found for NH_4^+, NO_3^-, and SO_4^{2-}, respectively. Since seasonality showed a pronounced year-to-year variability in the ice core, it is unclear whether these 1-year data indicate a significant difference between air and snow concentrations. However, they are in line with results from several years' aerosol monitoring at the Jungfraujoch (3450 m asl, Swiss Alps), and the Colle Gnifetti (4450 m asl, Swiss Alps), where comparable high summer to winter ratios of the aerosol surface area concentration were detected [*Lugauer et al.*, 1998]. A difference in the extent of the seasonality of concentrations in air and snow might be due to a seasonal variation of the scavenging ratios with higher values in the cold and lower values in the warm season [*Wagenbach*, 1997].

Concentrations of Cl^-, Ca^{2+}, K^+, and Na^+, on the other hand, fluctuate more episodically and show nearly no seasonality (summer to winter ratios between 1.03 and 1.33, Table 2). High concentrations of Ca^{2+} are attributed to long-range transport events of Saharan dust, contributing substantially to the annual deposition of Ca^{2+} and carbonate [*Schwikowski et al.*, 1995]. Saharan dust transports toward the Alps occur episodically with a slightly higher frequency from early spring to summer [*Prodi and Fea*, 1995]. However, it appears from our record that the main events are nearly equally distributed between winter and summer snow. Besides the Saharan dust induced concentration peaks, the background concentrations of Ca^{2+} fluctuate without any seasonal preference. A similar behavior is observed for background concentrations of Cl^-, K^+, and Na^+, while the maximum concentrations in this case point to a contribution of sea spray (see above).

4.3. Concentration Trends

In order to extract the trends from the concentration records, annual averages were calculated for the various species. For this purpose, the segments of the core covering a year were defined using the $\delta^{18}O$ signal, with the minima serving as separating parameter. However, one has to bear in mind that the minimum $\delta^{18}O$ occurs normally in February. The resulting concentration records still show a strong year-to-year variation of the average values (Figures 6a and 6b). Nevertheless, for concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ an increasing trend is revealed from 1946 until about 1975, reflecting the growing anthropogenic emissions of the precursor gases. After reaching the maximum, concentrations of the three species declined, a trend which is most pronounced for SO_4^{2-}. This decline is due to reduced anthropogenic emissions [*Mylona*, 1993]. A different behavior was found for Cl^- and Na^+, where concentrations showed a constant level until 1960, increased during a 5-year period, and remained constant again at a higher level for the rest of the time. This suggests a change in the frequency or strength of the sea-salt-transporting weather conditions, which is consistent with an increase in the number of days with westerly winds in Europe starting about 1950 (H. Wanner, unpublished data, 1997). A significant additional source of chlorine, e.g., HCl from incineration, can be excluded, since it would have enhanced only the concentration of Cl^-. In contrast, no significant concentration trends were observed for Ca^{2+}, K^+, and Mg^{2+}. However, they show even stronger year-to-year variations than the other species, and it is therefore not possible to identify trends if they are not strong. This result contradicts a long-term Alpine mineral dust record from Colle Gnifetti, exhibiting an increase of background Ca^{2+} (defined as including anthropogenic pollution, but no dusts events) since the second half of the 19th century [*Wagenbach et al.*, 1996]. For $\delta^{18}O$ no trend could be seen (Figure 6b), indicating that the observed concentration trends were not caused by varying precipitation characteristics but, indeed, represent changing emissions and ambient air conditions. This is principally corroborated by the record of the layer thickness (Figure 6b), although it showed lower values in the deepest part of the core. They are likely due to layer thinning; however, it was

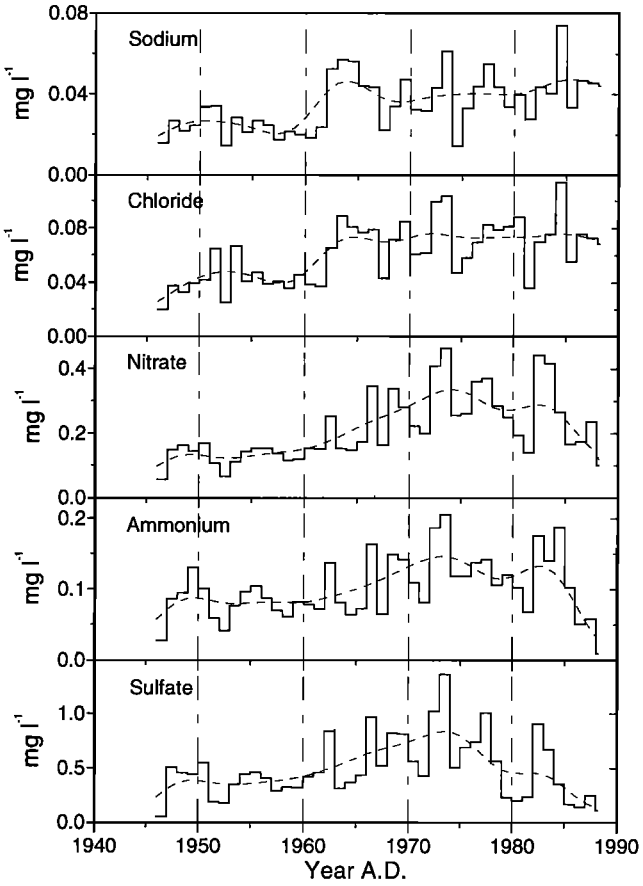

Figure 6a. Concentration records of SO_4^{2-}, NH_4^+, NO_3^-, Cl^-, and Na^+ (annual averages). Dashed curves represent smoothed values to guide the eye.

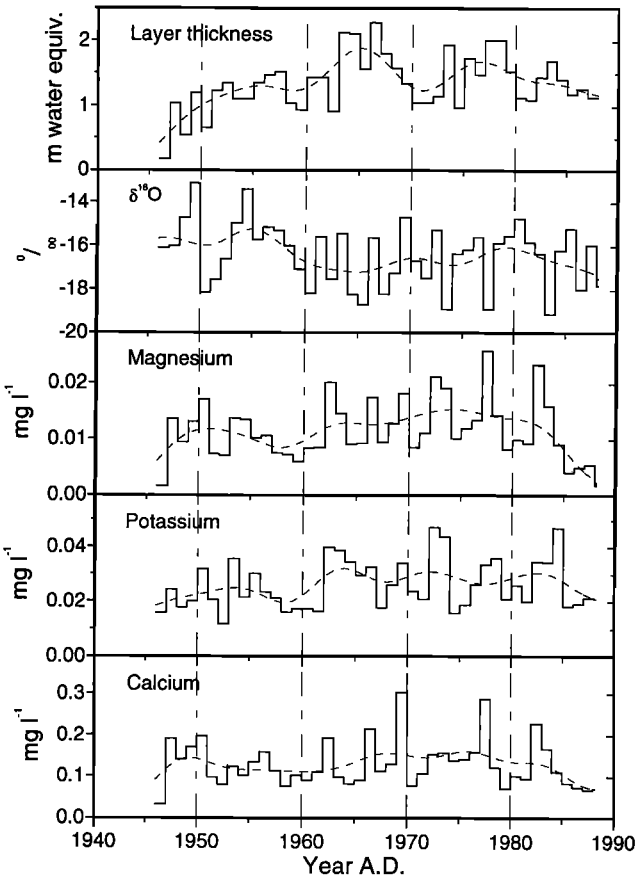

Figure 6b. Same as Figure 6a, but for Ca^{2+}, K^+, and Mg^{2+} as well as $\delta^{18}O$, along with the corresponding annual layer thickness.

not possible to correct for this effect, since no glaciological models exist for glaciers with a limited horizontal and vertical extension located on a slope.

As explained above, winter months at high-alpine sites are characterized by a decoupling from the underlying atmospheric boundary layer. Thus it is assumed that free tropospheric conditions prevail. In order to investigate concentration trends in snow representing free tropospheric conditions, the classification in winter and summer snow based on $\delta^{18}O$ was used; that is winter snow was defined as snow with $\delta^{18}O \leq -19^0/_{00}$, and summer snow was defined as snow with $\delta^{18}O > -15^0/_{00}$. The resulting summer and winter concentration records for SO_4^{2-} are illustrated in Figure 6c. The trend of the summer concentrations is similar to that obtained when considering all data, whereas the winter record is dominated by a few years with exceptionally high annual values (1966, 1969, 1972, 1973, and 1982). This indicates that atmospheric transport to the high-alpine site might occur even under temperature conditions typical for winter and reveals the limits of the applied classification scheme where a temperature proxy ($\delta^{18}O$) was used to discriminate between different transport conditions.

4.4. Calibration of the Paleo Record With Atmospheric Measurements

Since 1973, aerosol SO_4^{2-} concentrations have been determined with a time resolution of 48 hours at the high-alpine research station Jungfraujoch [*Gehrig*, 1996]. The Jungfraujoch is located about 6 km west of the Fiescherhorn glacier at an elevation of 3450 m asl, and we assume that atmospheric concentrations are comparable at both sites owing to the small horizontal and vertical distances. The aerosol SO_4^{2-} concentration record illustrated in Figure 7 shows the seasonal variation, with a

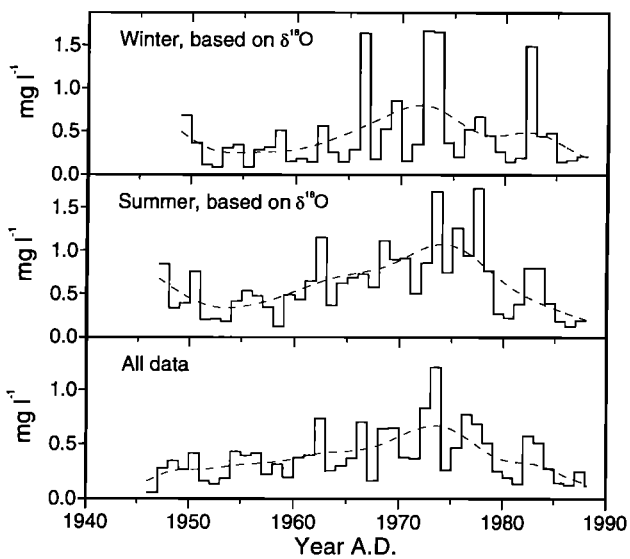

Figure 6c. Concentration records of SO_4^{2-}, classified in summer and winter snow (annual averages), based on the $\delta^{18}O$ values. For comparison the record consisting of all data is shown, too. Dashed curves represent smoothed values to guide the eye.

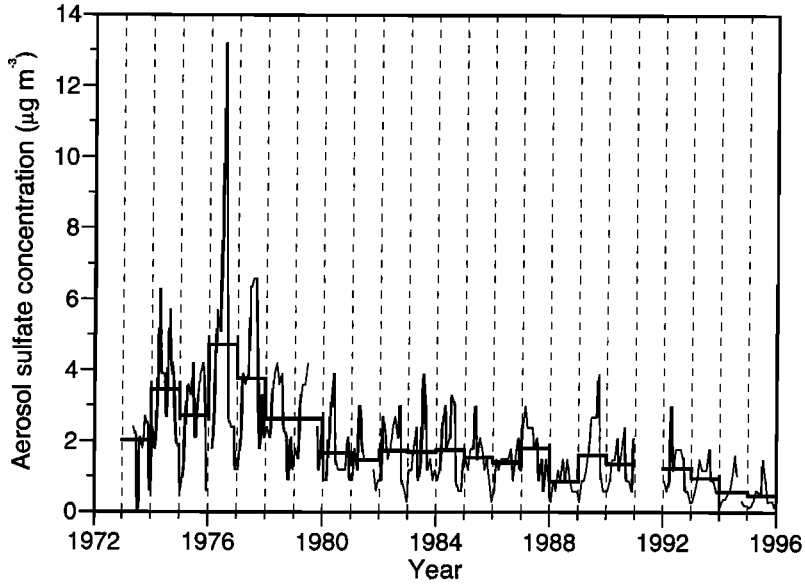

Figure 7. Aerosol SO_4^{2-} concentration at the high-alpine site Jungfraujoch (3450 m above sea level). Thin solid line represents monthly averages, and bold solid line denotes annual averages. Data are from *Gehrig* [1996].

maximum in summer and a minimum in winter, typical for a high-elevation mountain site as explained above. In order to calibrate the Fiescherhorn SO_4^{2-} ice record with the atmospheric data, annual averages were calculated for both over the time period with complete annual data sets (1974-1988). A higher time resolution could not be deduced from the paleorecord, since precipitation occurs rather episodically, making exact timing impossible. Both records correlate well, as illustrated in Figure 8 ($[SO_4^{2-}{}_{(firn/ice)}]$ = 180 $[SO_4^{2-}{}_{(air)}]$, r^2=0.41, concentrations in μg kg^{-1}). The concentrations of SO_4^{2-} in the atmosphere are thus indeed preserved in the ice core. Although no long-term atmospheric measurements exist for the other species investigated in this study, it can be assumed that their concentrations in the air are equally well recorded in the glacier ice, since as aerosol-borne constituents, they are expected to show similar behavior. From the linear regression analysis a mass scavenging ratio W of 180 for SO_4^{2-} (range for individual years; 89-455; average; 186±111) was obtained ($W = \rho_a C_s/C_a$, where C_s is concentration in snow in μg L^{-1}, C_a is concentration in air in μg m^{-3}, and ρ_a is density of air with a value of 0.875 g L^{-1} for the conditions at the Jungfraujoch). This value is in good agreement with scavenging ratios for SO_4^{2-} deposited on the Greenland ice sheet at Dye 3 (180±120, three summer snowfalls [*Davidson et al.*, 1985]), at Dye 3 calculated using atmospheric concentrations measured at coastal sites (100-200 in winter, 200-400 in summer [*Davidson et al.*, 1987]), and at Summit, Greenland (220±129, eight summer snowfalls [*Davidson et al.*, 1996]). A higher scavenging ratio of 350 was reported from simultaneous aerosol and snow sampling at the Alpine site Weissfluhjoch Davos, Switzerland, for a 3-month sampling period [*Baltensperger et al.*, 1993], whereas values from single event studies at the two high-alpine sites Jungfraujoch, Switzerland, and Sonnblick, Austria, are significantly higher (710 [*Schwikowski et al.*, 1995], 880 and 1500 [*Wagenbach*, 1997]). Comparably high scavenging ratios were determined for nonsea salt SO_4^{2-} at Antarctic stations (960 and 940 at Neumayer; 3270 at Halley [*Wolff et al.*, 1998]). The variety of reported data possibly illustrates the complexity of the scavenging process. However, it seems that event-based studies reveal higher values, probably because they reflect only the ambient conditions during synoptic situations with precipitation. Hence it is particularly important for the interpretation of paleorecords to establish the relationship between atmospheric and paleodata for the respective conditions at the glacier site.

The comparison of atmospheric and ice core concentrations reveals another important feature of the Fiescherhorn paleo atmospheric record, the pronounced year-to-year variability (Figure 8), which is not caused by the much more smooth atmospheric concentrations. Thus the ice core signal seems to be additionally modulated by climatic variations, e.g., variations of time of the year when most of the precipitation occurs, and atmospheric concentration or emission changes can therefore only be deduced when considering time periods significantly longer than 1 year.

5. Conclusions

The Fiescherhorn glacier is a site in the north of the main Alpine chain that meets all the requirements of an environmental archive. The part of the glacier where the drilling was performed belongs to the cold infiltration zone, thus disturbance of the chemical stratigraphy can be assumed to be negligible. A high net accumulation of, on average, 1.4 m water equivalent per year allowed, on one hand, the dating of the ice core by annual layer counting of the seasonally varying signals of the tritium concentration and of the isotopic ratio $\delta^{18}O$, revealing the time period 1946-1988 covered by the ice core. On the other hand, high-resolution chemical concentration records were accessible, showing that the snow composition at Fiescherhorn is dominated by constituents originating from anthropogenically derived secondary aerosol particles such as SO_4^{2-}, NH_4^+, and NO_3^-, as well as by mineral dust components such as Ca^{2+}. This composition characterizes the Fiescherhorn site as a relatively unpolluted continental site, in agreement with results from other

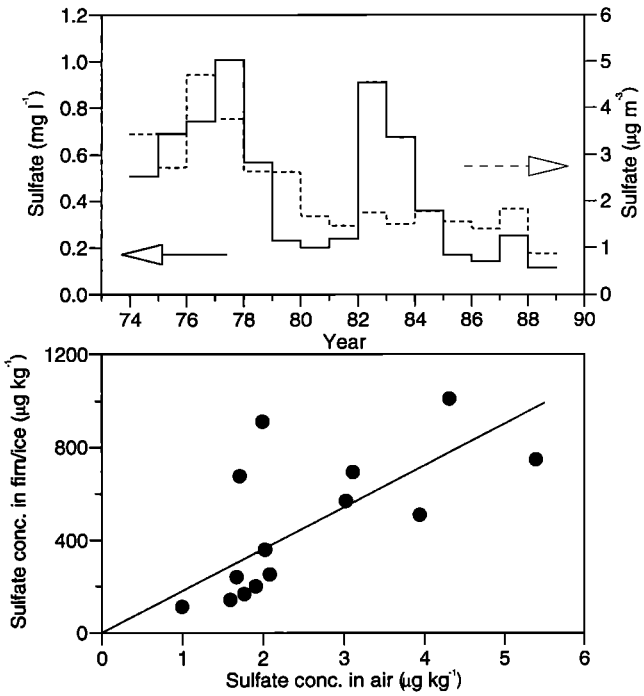

Figure 8. (top) Annual SO_4^{2-} averages determined in the Fiescherhorn ice core (solid line) and in atmospheric samples collected at the nearby high-alpine site Jungfraujoch (dashed line, data from *Gehrig* [1996]) for the time period 1974-1988. (bottom) Scatterplot of the annual averages, along with the resulting linear regression line ($[SO_4^{2-}_{(firn/ice)}] = 180\ [SO_4^{2-}_{(air)}]$, correlation coefficient r^2=0.41). For the regression the intercept was forced to intersect the origin, yielding a standard error of the slope of 22 and a p value of $2x10^{-6}$ (n=14, 1979 excluded because of incomplete atmospheric data).

cold glaciers in the Alps (Colle Gnifetti, Monte Rosa, and Col du Dome, Mont Blanc).

In the case of the secondary aerosol-borne species SO_4^{2-}, NH_4^+, and NO_3^-, a pronounced seasonality of the concentrations with higher values in summer and lower values in winter was observed. This is in line with results from aerosol monitoring at high-alpine sites, showing that the seasonality of the atmospheric stability combined with a strong vertical concentration profile is causing the seasonality of the aerosol concentration.

Concentration trends were extracted from the Fiescherhorn records by calculating annual averages, revealing an increasing trend for the species SO_4^{2-}, NH_4^+, and NO_3^- from 1946 until about 1975 and, after reaching this maximum, a decreasing trend until 1988. Thus the ice core indeed preserves the changing concentrations in the atmosphere that are due to changing emissions. For the mineral dust tracers, no trend was obvious, whereas Cl^- and Na^+ concentrations showed a slightly higher level from 1965 until 1988, indicating a change in the frequency or strength of sea salt transport. A significant anthropogenic input of HCl could not be detected.

A comparison of the SO_4^{2-} concentration record from Fiescherhorn with a direct atmospheric record from the nearby Jungfraujoch site for the time period 1974-1988 showed good agreement ($[SO_4^{2-}_{(firn/ice)}] = 180\ [SO_4^{2-}_{(air)}]$, r^2=0.41). Thus the Fiescherhorn paleorecord is unique, since it could be "calibrated" over a significant period of time, revealing a mean mass scavenging ratio of 180 for SO_4^{2-}.

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